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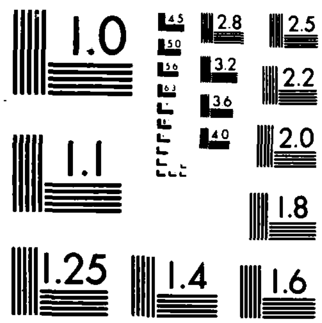

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Laser Spectroscopy, Optical Collisions, Radiative Collisions, Two-level Atom, Atomic Coherence, Degenerate Four-Wave Mixing, Saturation Spectroscopy, Laser Propagation, Dressed Atom, Photon Echo, Bloch Equations, Collision Kernel, Collisions, Optical Noise

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Work is reported in areas of:-
(1) Creation of Electronic State Coherences via Laser-Assisted Collisions
(2) Collision Effects in Four-Wave Mixing and Pump-Probe Spectroscopy
(3) Modified Optical Bloch Equations in Solids
(4) Transient Spectra in Atom-Field Interactions
(5) Collision Kernels and Transport Coefficients

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(6) Collisions in Strong cw and Transient Laser Fields

(7) Quantum vs Classical Description of Laser Fields

(8) Intense Optical Noise Sources

(9) Laser-Assisted Collisions

Research has been carried out in the areas of (1) Modified Optical Bloch Equations in vapors and solids, (2) Creation of electronic-state coherence in laser-assisted collisions (3) Collisionally modified line shapes in pump-probe and 4-wave mixing spectroscopy, (4) Collision kernels and transport coefficients, (5) Collisions in strong cw and transient laser fields, (6) Quantum vs classical description of laser fields in nonlinear spectroscopy, (7) Intense optical noise sources, (8) Laser-assisted collisions and (9) Transient spectra in atom-field interactions.

1. Modified Optical Bloch Equations (P. Berman)

The work outlined in last year's annual report[1] has been completed. A "strong redistribution" model of Markovian fluctuations was used to analyze the Free-Induction Decay (FID) signal emitted by atoms which were subjected to random frequency fluctuations.[2] The results of this calculation helped to explain in a qualitative manner the experimental results of DeVoe and Brewer[3] which had stimulated a great deal of theoretical activity in this area. A detailed examination of the validity requirements for the Optical Bloch Equations was presented in a second article.[4*] In a companion article,[5*] a general method was developed for calculating the effects of atomic frequency fluctuations in laser-atom interactions. This method is particularly useful if the atom-field interaction can be treated in a perturbative expansion. Explicit time-dependent and time-independent frequency fluctuation propagators were derived for (a) strong redistribution, (b) "difference kernel" and (c) Brownian motion models for the frequency fluctuations. It was shown that no consistent explanation of the DeVoe-Brewer results seems to exist. The next step in this problem would be to extend the method to include saturation effects; this extension represents an extremely difficult task.

2. Creation of Electronic-State Coherence in Collisionally-Assisted Collisions (P. Berman)

Experimental evidence has been obtained for the creation of collision-

* An asterisk indicates that a reprint or preprint of this article has been forwarded to the Scientific Officer with this report. Reprints of articles have been furnished to DTIC with this report. Preprints or reprints of these articles are available on request to anyone receiving this report.

induced, optical coherence in Na.[6*] The reaction that was studied is

$$Na(3S_{1/2}) + He + \hbar\Omega_1 + 2\hbar\Omega_2 \rightarrow Na(4D\text{-}3P_{3/2}) + He$$

where the notation $(4D\text{-}3P_{3/2})$ indicates a coherent superposition of the 4D and $3P_{3/2}$ states. The laser at frequency Ω_1 is tuned about 200 GHz below the $3S\text{-}3P_{3/2}$ transition frequency. When the second laser's frequency Ω_2 is scanned, a resonance structure is observed when $2\Omega_2$ is detuned from the 3S-4D transition by the same amounts as the first laser. This resonance structure is observed in emission at the $4D\text{-}3P_{3/2}$ transition frequency (essentially via a 4-wave mixing process), but is absent in the absence of collisions. Thus, it has been demonstrated that it is possible to generate coherent optical radiation in a collision-induced reaction.

3. Collisionally-Modified Line Shapes in Pump-Probe and 4-wave Mixing Spectroscopy (G. Khitrova, P.R. Berman)

G. Khitrova has finished her thesis in which new features of the line shapes in pump-probe spectroscopy and 4-wave mixing on a single atomic transition are examined. In particular, new effects related to (a) effects of magnetic degeneracy, (b) velocity-changing collisions and (c) contributions from all velocity subclasses of atoms (and not from only those Doppler-shifted into resonance with the fields) have been reported. The conditions under which narrow resonances (characterized by the ground state width) can be observed are clearly established. A short summary of the various topics is contained in last year's Annual Report.[1]

4. Collision Kernels and Transport Coefficients (P. Berman)

In collaboration with the group of J. Woerdman (University of Leiden), we have established an integral relation between the collision kernels used in analyzing laser spectroscopy experiments and the transport coefficients of classical transport theory.[8*] Our results are extremely useful in the analysis of laser spectroscopy experiments involving collisions. By using experimental or theoretical values for the various transport coefficients, one can reduce the number of free parameters needed to fit laser spectroscopic line shapes in which collisions play a role. This method has been applied to a study of Na - rare gas collisions, observed using saturation spectroscopy. Good fits to the data were obtained with essentially no free parameter. A knowledge of collisional parameters for the Na - rare gas system is critical for understanding the theory of the so-called "optical piston".[9]

5. Collisions in Strong cw and Transient Laser Fields (P. Berman)

Attempts continue to obtain "propagators" which give the time-development for a "two-level" atom subject to both a strong external field and atomic phase and frequency fluctuations. The original motivation for this problem is linked to an experiment[10] in Mossberg's group in which he claimed quenching of the effects of velocity-changing collisions at high laser field strengths in a photon echo experiment. As yet, there has been no rigorous proof of that conjecture. Our calculation is intended to supply an answer to this problem. Some progress has been made using a dressed-atom approach to the problem, but we are still unable to confirm or reject Mossberg's interpretation of his result. The problem we are studying is fundamental and the results, when obtained, can be applied to a wide variety of problems involving atoms in strong fields undergoing phase or frequency fluctuations.

6. Quantum vs Classical Descriptions of Laser Fields in Nonlinear Spectroscopy (P. Berman)

In collaboration with G. Grynberg , we have started to examine some aspects of nonlinear spectroscopy related to the quantized nature of the electromagnetic field. The motivation for this study is based on obtaining a physical understanding of "collision-induced extra resonances" in 4-wave mixing experiments.[11] In such experiments, it is often stated that collisions lead to a "real" (i.e. non-virtual) excitation of level coherence. If the level coherence in such a problem is calculated using a fixed number quantized radiation field, it can be shown that the level coherence vanishes identically. Does this mean that the pressure-induced resonance

resonance would vanish if fixed number states of the radiation field were used experimentally? To answer this question, we have begun a systematic study of various experimental situations of current interest to determine which of these experiments would provide the most sensitive probe of the quantum vs classical nature of the radiation field.

7. Intense Optical Noise Sources

Several experimental groups[12] are now using large-band laser sources as a means for obtaining femtosecond time resolution. The major idea behind the work is that one can exploit the short coherence time (τ_c = 1.0 ps) of a large-band source to arrive at a time resolution which is of order τ_c. Stimulated photon echo experiments, have been carried out which have dramatically demonstrated the feasibility of this idea.[12] However, there have been field saturation effects which cannot be explained within the context of current theories. We have started to formulate a theory for the stimulated photon echo with broad-band noise sources. Some progress has been made analytically, but a large scale numerical solution may be necessary. The theoretical problem is complicated by the fact that the same noise is sent into an atomic vapor with a time delay. This means that the system has a memory and this memory greatly complicates the analysis.

8. Laser-assisted Collisions (P. Berman)

In collaboration with A. Bambini (Quantum Electronics Institute, Florence), we have tried to explain a long-standing mystery regarding the far wing behavior of LICET (laser-induced collisional energy transfer) profiles. Theory, based on an effective two-level problem, predicts a $\Delta^{-0.5}$ (Δ = detuning from resonance) dependence while experiment varies from $\Delta^{-0.5}$ to $\Delta^{-0.85}$. Improving the existing calculations by including some additional contributions from a third (virtual) level has enabled us to give a unified explanation of existing experiments. An article is in preparation.

In collaboration with F. Schuller (High-Pressure Laboratory - Villetaneuse, France) we are trying to extend a previous calculation[13] on effects of magnetic degeneracy in LICET into the line wing.

9. Transient Spectra in Atom-Field Interactions

In collaboration with the group of T. Mossberg (Harvard), we have studied the transient spectra produced when atoms are prepared in pure dressed states of the atom and radiation field. Both transient Autler-Townes spectra[14*] and transient resonance fluorescence[15*] for atoms prepared in dressed states have been studied theoretically. It was shown that one of the peaks in the Autler-Townes doublet and fluorescence triplet is suppressed at early times. A simple explanation, based on a dressed-atom picture, was used to understand the results. Experimental verification of the transient Autler-Townes doublet for pure dressed-state preparation has been achieved.[16*] Theoretical calculations for the transient pump-probe spectroscopy of a single transition is in progress.

10. Miscellaneous

A previous review article on optical pumping in nonlinear spectroscopy has appeared as has our work on photon echoes using double-resonance pulses.[17*,18*] Previous works on two-level systems[19*,20*] corrections to exponential decay produced by adiabatic switching[21*] and a rederivation of Wigner's infinite product representation of the R matrix[22*] were published. The effect of an unstable continuum on the exponential decay law was studied.[23*] A calculation relating to the treatment of intermediate states in constructing Green function operators was carried out.[24*] An analogy between certain integrals appearing in the theory of resonance fluorescence following pulse excitation and certain diffraction integrals was obtained.[25*] Finally, some earlier work on pulse propagation involving the interaction of two fields with a "three-level" atom has appeared.[26*]

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2. P.R. Berman, J.E.M. Haverkort and J.P. Woerdman, "Collision Kernels and Transport Coefficients," Physical Review A (to appear) (additional support from NSF).
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